CLAIMS

1. The use of a starting solution to form a plurality of filler elements in a composite material, wherein the starting solution is a miscible solution of at least one protein and a least one further compound and whereby the plurality of filler elements are formed from the at least one further compound.

2. The use of claim 1 in which the plurality of filler elements are filler particles, filler fibrils, elongate fluid-filled cavities, or micelles.

3. The use of claim 1 or 2, wherein the starting solution contains a mixture of two or more proteins or synthetic protein analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins.

4. The use of claims 1 to 3 wherein the starting solution comprises a phase separating agent selected from the group of phase separating agents consting of proteoglycans, glycosaminoglycans, carbohydrates (such as trehalose or sucrose), polyols, peptides or proteins rich in serine and or threonine, glycerol and its derivatives, and detergents.

5. A method of forming a composite material containing a plurality of filler elements comprising:
i) a first step of preparing a starting solution containing a miscible solution of at least one protein and at least one further compound; and
ii) a second step of inducing the starting solution to separate into a bulk phase and a minor phase.

6. The method of claim 5, wherein the starting solution comprises two or more proteins or synthetic protein analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins.

7. The method according to any one of claim 5 or 6, wherein the starting solution comprises a phase separating agent selected from the group of phase separating agents consisting of proteoglycans, glycosaminoglycans, carbohydrates (including trehalose and sucrose), polyols, peptides or proteins rich in serine and or

threonine, glycerol and its derivatives, and detergents

8. The method according to one of claim 5 to 7 wherein the starting solution additionally contains a solvent and the second step is initiated by withdrawing the solvent from the starting mixture.

9. The method of claim 8 wherein the withdrawal of the solvent is carried out by drying the starting mixture, by the addition of a solvent-binding agent or agents, by altering the pH, by altering the temperature, by changing the pressure by adding inorganic salts, by adding other phase separating compound or compounds, or by a combination of two or more of these factors.

10. The method according to any one of claims 5 to 9, wherein the starting solution comprises a mixture of spidroin I and spidroin II or analogues thereof.

11. A method according to any one of claims 5 to 9, wherein the starting solution contains a mixture of fibroin proteins or analogues thereof.

12. The method according to any one of claims 5 to 9, wherein the starting solution comprises fibrous proteins.

13. The method according to any one of claims 10 to 12, wherein the starting solution comprises native or recombinant-produced proteins or protein fragments.

14. A method according to any one of claims 5 to 9, wherein the at least one protein of the starting solution comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

15. The method according to any one of claims 5 to 14, wherein the minor phase comprises micelles.

16. The method according to any one of claims 5 to 14, wherein the minor phase comprises small droplets.

17. The method according to claim 16 wherein the small droplets are fluid droplets.

18. The method according to claim 16 wherein the small droplets are caused to solidify to become solid filler particles.

19. The method according to claim 16 wherein the minor phase includes at least one material, which becomes cross-linked to form either hard or elastomeric filler elements.

20. The method according to claim 16 wherein the small droplets are caused to elongate into elongated filler fibrils or elongated fluid filled cavities.

21. The method of any one of claims 5 to 20 further comprising a step of flowing the composite material through a die.

22. The method of claim 21, wherein the die has a convergent form with a converging surface along and towards the axis of the die.

23. The method of claim 21 or 22, wherein the minor phase is elongated substantially parallel to the parallel axis of the die.

24. The method according to claim 21 wherein the die has a divergent form.

25. The method according to claim 24 wherein the small droplets are caused to elongate into elongated filler fibrils or elongated fluid filled cavities orientated in curved hoops describing the axes of elongation as the composite material is flowed through the die

26. A method according to any one of claims 21 to 25, wherein the minor phase remains fluid as the composite material is flowed through the die to form elongated fluid filled cavities.

27. The method according to any one of claims 21 to 25, wherein the composite material is initially present as a fluid within which the minor phase solidifies

during passage through the die to form elongated solid fibrils.

28. The method according to any one of claims 21 to 25, wherein the composite material is initially present as a fluid within which the minor phase is cross-linked during passage through the die to form elongated hard or elastomeric fibrils.

29. A method of extruding a starting material to form a composite material containing a plurality of filler elements comprising:
i) a first step of preparing the starting solution;
ii) a second step of inducing the starting solution to separate into a bulk phase and a minor phase; and
iii) a third step of extruding the starting material either prior to or coincident with the second step to form the composite material.

30. The method of claim 29, wherein the starting solution comprises two or more proteins or synthetic protein analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins.

31. The method according to any one of claims 28 or 29, wherein the starting solution comprises a phase separating agent selected from the group of phase separating agent consisting of proteoglycans, glycosaminoglycans, carbohydrates such as trehalose, sucrose, polyols, peptides or proteins rich in serine and or threonine, glycerol and its derivatives, and detergents.

32. The method according to any one of claims 29 or 31, wherein the starting solution additionally contains a solvent and the second step is initiated by withdrawing the solvent from the starting mixture.

33. The method of claim 32, wherein the withdrawal of the solvent is carried out by drying the starting mixture, by the addition of a solvent-binding agent or agents, by altering the pH, by altering the temperature, by changing the pressure, by adding inorganic salts, by adding other phase separating compound or compounds, or by a combination of two or more of these factors.

34. The method according to any one of claims 30 to 33, wherein the starting solution comprises a mixture of spidroin I and spidroin II or analogues thereof.

35. A method according to any one of claims 30 to 33, wherein the starting solution contains a mixture of fibroin proteins or analogues thereof.

36. The method according to any one of claims 30 to 33, wherein the starting solution comprises fibrous proteins.

37. The method according to any one of claims 34 to 36, wherein the starting solution comprises native or recombinant produced proteins or protein fragments.

38. A method according to any one of claims 30 to 33, wherein the at least one protein of the starting solution comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

39. The method according to any one of claims 29 to 38, wherein the minor phase comprises small droplets.

40. The method according to any one of claim 29 to 38, wherein the minor phase comprises small droplets.

41. The method according to claim 40, wherein the small droplets are fluid droplets.

42. The method according to claim 40, wherein the small droplets are caused to solidify to become solid filler particles.

43. The method according to claim 40, wherein the minor phase includes at least one material, which becomes cross-linked to form either hard or elastomeric filler elements.

44. The method according to claim 40, wherein the small droplets are caused to elongate into elongated filler fibrils or elongated fluid filled cavities.

45. The method of any one of claims 29 to 44 further comprising a step of flowing the composite material through a die.

46. The method of claim 45, wherein the die has a convergent form with a converging surface along and toward a parallel axis of the die.

47. The method of claim 45 or 46, wherein the minor phase is elongated substantially parallel to the parallel axis of the die.

48. The method according to claim 45 wherein the die has a divergent form.

49. The method according to claim 48, wherein the small droplets are caused to elongate into elongated filler fibrils or elongated fluid filled cavities orientated in curved hoops describing the axes of elongation as the composite material is flowed through the die

50. A method according to any one of claims 45 to 49, wherein the small droplet remains fluid as the composite material is flowed through the die to form elongated fluid filled cavities.

51. The method according to any one of claims 40 to 50, wherein the composite material is initially present as a fluid within which the minor phase solidifies during passage through the die to form elongated solid fibrils.

52. The method according to any one of claims 45 to 50 wherein the composite material is initially present as a fluid within which the minor phase is cross-linked during passage through the die to form elongated hard or elastomeric fibrils.

53. An apparatus for forming a composite material from a starting solution having:
- a storage compartment for storing the starting mixture containing a miscible solution of at least one protein and at least one further compound; and
- a phase separation compartment for separating the at least one protein and the at least one further compound of the phase separating mixture into a bulk material containing a plurality of filler elements.

54. The apparatus of claim 53, wherein the filler elements are particles, filler fibrils, elongated fluid filled cavities or micelles.

55. The apparatus according to claim 53 or 54, wherein the phase separation compartment comprises a compartment for withdrawing a solvent from the starting mixture by drying, by the addition of a solvent-binding agent or agents, by altering the pH of the starting mixture, by altering the temperature, by changing the pressure, by adding inorganic salts to the starting mixture, by adding to the starting mixture other phase separating compound or compounds or a combination of these.

56. The apparatus according to claim 53 or 54, wherein the phase separation compartment comprises a compartment in which the small droplets are caused to elongate.

57. The apparatus according to claim 53 or 54 wherein the phase separation compartment comprises a compartment in which the filler elements are allowed to solidify or become cross-linked.

58. The apparatus according to any one of claims 53 to 57, wherein the starting mixture contains a mixture of two or more native proteins or synthetic protein analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins

59. The apparatus according to any one of claims 53 to 58, wherein the starting solution comprises a mixture of spidroin I and spidroin II or analogues thereof.

60. The apparatus according to any one of claims 53 to 57, wherein the starting solution contains a mixture of fibroin proteins or analogues thereof.

61. The method according to any one of claims 53 to 57, wherein the starting solution comprises fibrous proteins.

62. The method according to any one of claims 53 to 57, wherein the starting solution comprises native or recombinant-produced proteins or protein fragments.

63. The apparatus according to any one of claims 53 to 57, wherein the starting solution comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

64. An apparatus for extruding a composite material from a starting solution having:
- a storage compartment for storing the starting mixture;
- a phase separation compartment for separating the starting mixture into a bulk material containing a plurality of filler elements; and
- an extrusion compartment for extruding the bulk material.

65. The apparatus of claim 64, wherein the plurality of filler elements are particles, filler fibrils, elongated fluid filled cavities or micelles.

66. The apparatus according to claim 64 or 65, wherein the phase separation compartment comprises a compartment for withdrawing a solvent from the starting mixture by drying, by the addition of a solvent-binding agent or agents, by altering the pH of the starting mixture by altering the temperature, by changing the pressure, by adding inorganic salts to the starting mixture, by adding to the starting mixture other phase separating compound or compounds or a combination of these.

67. The apparatus according to claim 64 or 65 wherein the phase separation compartment comprises a compartment in which the small droplets are caused to elongate.

68. The apparatus according to claim 64 or 65 wherein the phase separation compartment comprises a compartment in which the plurality of filler elements are allowed to solidify or become cross-linked.

69. The apparatus according to any one of claims 64 to 68, wherein the starting mixture contains a mixture of two or more proteins or synthetic protein

analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins

70. The apparatus according to any one of claims 64 to 69, wherein the starting solution comprises a mixture of spidroin I and spidroin II or analogues thereof.

71. The apparatus according to any one of claims 64 to 69, wherein the starting solution contains a mixture of fibroin proteins or analogues thereof.

72. The apparatus according to any one of claims 64 to 69, wherein the starting solution comprises fibrous proteins.

73. The apparatus according to any one of claims 64 to 69, wherein the starting solution comprises native or recombinant-produced protein fragments.

74. The apparatus according to any one of claims 64 to 69, wherein the starting solution comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

75. Composite material made according to the method of any one of claims 5 to 52.

76. Composite material comprising at least one protein and at least one further compound, wherein the composite material has a bulk phase and a plurality of filler elements and wherein at least one of the bulk phase or the plurality of filler elements comprise the at least one protein.

77. The material of claim 76, wherein the composite material comprises two or more proteins or synthetic protein analogues or synthetic polymers or a mixture of synthetic polymer or polymers and one or more proteins

78. The material of claim 76 or 77, wherein the composite material comprises a mixture of spidroin I and spidroin II or analogues thereof.

79. The material of claim 76 or 77, wherein the composite material contains a mixture of fibroin proteins or analogues thereof.

80. The material of claim 76 or 77, wherein the composite material contains fibrous proteins.

81. The material of claim 76 or 77, wherein the composite material contains native or recombinant-produced proteins or protein fragments.

82. The material of claim 76, wherein the at least one protein of the composite material comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

83. Starting material for use in the preparation of a composite material with a plurality of filler elements comprising a miscible solution of:
at least one protein; and
at least one other compound, whereby at least partial phase separation between the at least one protein and the at least one other compound can be induced to form composite material with the plurality of filler elements.

84. The material according to claim 83 further comprising a solvent and wherein the at least partial phase separation occurs by removal of the solvent.

85. The material according to claim 83 or 84, further comprising a mixture of two or more proteins, or synthetic protein analogues, synthetic polymers, or polymers and one or more proteins, or fragments thereof.

86. The material according to one of claims 83 to 85 comprising a mixture of spidroin I and spidroin II or analogues thereof.

87. The material according to one of claims 83 to 85 comprising a mixture of fibroin proteins or analogues thereof.

88. The material according to one of claims 83 to 85 comprising fibrous proteins.

89. The material according to one of claims 83 to 85 comprising native or recombinant-produced proteins or protein fragments.

90. The material according to claims 83 to 85, wherein the at least one protein of the starting solution comprises a spinning dope extracted from the silk gland of a lepidopteran insect or from an arachnid.

91. Composition according to any one of claims 84 to 90, wherein the solvent comprises at least water.